From nano- to global scales: properties, processes of formation, and aftereffects of atmospheric aerosol impacts. 6. Long-range transport and settling processes

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The processes of long-range transport and settling of atmospheric aerosol are overviewed.

Introduction

The relatively long (up to 2–3 weeks) lifetime of aerosol particles in the atmosphere determines the possibility of their long-range transport. The best-known and most thoroughly studied situations are emissions of dust aerosol (DA) into the atmosphere during dust storms in the Northern Africa and the following transatlantic transport of particles, though sometimes meridional dust flows directed toward the Western Europe occur. Another no less known situation is the long-range transport of DA to the Northwestern Pacific Ocean during dust storm in the Northwestern China and in Mongolia (the brief review of the issue of dust storms can be found in Refs. 1 and 19).

The realization of the key role of aerosol in formation of climate stimulated further development of the investigations into the long-range transport of aerosol.[1–50] This is evidenced, in particular, by the new ITCT-Lagrangian-2k4 program for studying the transcontinental transport and chemical transformation of aerosol.

Parrish and Law[35] characterized briefly the ITCT program, whose main goal is to understand the intercontinental transport and the chemical transformation processes of aerosols, oxidants and their precursors. To make this understanding possible, measurements from a Lagrangian platform would be ideal, i.e., a platform that moves with an air mass during the total transport process. Such an ideal is not possible, but a practical approximation to this ideal is a "pseudo-Lagrangian" study, where one or more aircraft make multiple, sequential sampling flights into the same air mass during the time required for the intercontinental transport of the air mass. Such a pseudo-Lagrangian study constitutes the ITCT task. The first stage of ITCT conducted in summer 2004 included airborne observations in the North Atlantic region, which focused on the study of emissions of aerosol and ozone precursors over North America, their chemical transformations and removal during transport to and over the North Atlantic, and their impact upon the Western Europe.

The key ITCT objectives are: 1) determination of the photochemical oxidant and aerosol formation potentials in polluted air masses originating in North American emission regions and their chemical evolution as they are transported out over the North Atlantic to the Western Europe; 2) characterization of the dynamical processes responsible for pollutant transport out of the North American planetary boundary layer; 3) quantification of the export of North American pollutants to the background atmosphere, their subsequent evolution and possible impact on climate. It is expected that more than 10 aircrafts from different countries will take part in the ITCT program.

To study regularities in intercontinental transport of atmospheric pollution, Stohl with co-workers[43] performed a 1-year simulation of the transport of six passive tracers, released over the continents according to an emission inventory for carbon monoxide (CO) (conditions of 2000 were considered). It was found that emissions from Asia experience the fastest vertical transport, whereas European emissions have the strongest tendency to remain in the lower troposphere. European emissions are transported primarily to the Arctic and appear to be the major contributor to the Arctic haze problem. Tracers from an upwind continent first arrive over a receptor continent in the upper troposphere, typically after some 4 days. Only later foreign tracers also arrive in the lower troposphere.

Assuming a 2-day lifetime, the domestic tracers dominate total columns over all continents except over Australia where foreign tracers account for 20% of the tracer mass. In contrast, for a 20-day lifetime even continents with high domestic emissions receive more than half of their tracer burden from foreign continents. Three special regions were identified where tracers are transported to, and tracer dilution is slow. Future field studies therefore should be deployed in the following regions: (1) In winter, the Asian tracer accumulates over Indonesia and Indian Ocean. (2) In summer, the highest concentrations of the Asian tracer are found in the Middle East. (3) In summer, the highest concentrations of the North American tracer are found in the Mediterranean.

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Predicting mineral aerosol distributions is a difficult task due to the episodic nature of the sources and transport. Based on 22-year satellite and *in situ* observations, Luo with co-workers[24] compared the observed spatiotemporal variability of the aerosol distribution with the results of numerical simulation obtained by joint use of the MATCH model of aerosol transport in the atmosphere with the allowance for chemical reactions, determining the transformation of its properties, and the DEAD model, reconstructing the processes of formation and transformation of dust aerosol.

On the whole the comparison revealed quite good agreement, but with some discrepancies. To analyze the reasons for these discrepancies, some tests of the sensitivity of mineral aerosol simulations to the meteorological data sets and schemes of mobilization parameterizations (mobilization here is inflow of particles into the atmosphere due to saltation) were conducted. The sensitivity analysis showed that near Australia the differences between the simulated spatial distribution of the aerosol optical depth (AOD) and the observed one were likely due to inadequate data about both the aerosol sources and the surface winds. Differences over East Asia are dominated by that the consideration of meteorology was quite unreliable.

According to the estimates obtained, the total emission of dust aerosol to the atmosphere as a result of dust storms is 1654 Tg/yr. The most powerful sources of DA emissions are African deserts, which contribute 73% to the total DA content in the atmosphere. The East Asia is a dominant contributor to formation of the dust aerosol field over the Pacific Ocean in the northern hemisphere, whereas Australia is the major source of DA in the southern hemisphere. The characteristic atmospheric lifetime of dust aerosol is about 6 days.

Important results characterizing the impact of aerosol on the atmosphere in the process of long-range transport were obtained by Meloni with co-workers.[30b] The PAUR II second observation phase was conducted over the island of Lampedusa, in the Mediterranean (35.5°N, 12.6°E), in May 1999, in order to study the photochemical processes in the atmosphere and the solar ultraviolet (UV) radiation. The ground-based instrumentation included a Brewer spectrophotometer, an aerosol lidar operated at 532 nm, a multi-filter rotating shadowband radiometer (MFRSR) to measure the net and diffuse radiation fluxes, as well as the aerosol optical depth at the wavelengths of 415, 500, 615, 671, 868, and 937 nm. Airborne measurements of actinic fluxes in the band of NO_2 photodissociation and in the band of O_3 photodissociation leading to the production of O(′D) were performed simultaneously.

Meloni et al.[30b] discussed the results obtained during three days: 18 May, when the outbreak of Saharan dust particles occurred in the atmosphere at 7 km altitude and a large aerosol optical depth (0.51 at 415 m) was measured; and 25 and 27 May, when low-to-moderate amounts of continental/marine aerosols (aerosol optical depth of about 0.2 at 415 m), originating from Europe, were present; the aerosol was confined below 3 km altitude in these 2 days. The measured actinic flux profiles were compared with the results obtained using radiative transfer model simulation; the model results were used to interpret the observations and study the sensitivity of the actinic flux profiles on the aerosol and ozone distributions. A good agreement between measurements and model results is obtained in all the cases examined.

Calculations at a fixed solar zenith angle showed that during the desert dust event the actinic flux in the band of NO_2 photodissociation was reduced to 24% in the lower and middle troposphere, with respect to the continental/marine aerosol cases. The actinic flux reduction increases with solar zenith angle, and the largest reduction occurs at the aerosol peak. The vertical distribution of aerosol particles did not influence the modeled fluxes at the surface, which basically depend on the aerosol column amount. When the climatological profile was used instead of the measured one, the actinic fluxes were within ±3% of the measurements on days when low-to-moderate optical depths occurred.

However, the modeled fluxes are largely overestimated, up to 19%, the measurements in the desert dust case. The analysis of the sensitivity of the actinic fluxes in the $O_3 + h\nu \rightarrow O_2 + O(′D)$ photodissociation region on the ozone distribution suggested that the ozone profile appears to affect the actinic fluxes in the O_3 photodissociation region, particularly at small solar zenith angles and near the surface. Because of an increase of the radiation scattering caused by aerosols in the troposphere, the model fluxes at the surface obtained with the climatological profile are 7.7% higher than those obtained with the measured profile for a zenith angle of about 37°; differences decrease with altitude and with the increase of the solar zenith angle.

1. African aerosol

Reid et al.[38] have analyzed the results of observation of the Saharan dust aerosol properties from the data of airborne and ground-based studies of the elemental composition of bulk DA and single particles. Airborne samples were collected on polycarbonate filters and underwent single-particle scanning electron microscopy with energy dispersive analysis with X-rays (EDAX). Particle chemistry was related to size and morphological characteristics. At the principal surface site, the Cabras Island (18.21°N, 65.60°W), particles were collected on a Davis Rotating Drum (DRUM) cascade impactor strips in eight stages from 0.1 to 12 μm (0.09, 0.24, 0.34, 0.56, 0.74, 1.1, 2.5, and 5 μm) at 4-hour time resolution. These samples were subjected to X-ray fluorescence (XRF) to determine bulk elemental composition from Al through Zn.

The data elements showed good correlation between the DRUM and the aircraft samples. Cluster analysis of single-particle data resulted in 63 statistically significant clusters. Several clusters can be easily related to their parent mineralogical species. However, as dust particles are to a large extent aggregates, most clusters are based on a continuum of varied mineralogical species and cannot be easily categorized. With 60500 total particles counted from the airborne filters, a statistically significant number of large particles could be analyzed. Estimated from the mean surface area modal diameter of particles was ~5 μm, with the average aspect ratio of 1.9. (Thus, the nonsphericity of the particle shape was quite pronounced.)

An apparent change in source region was seen in the morphological data and non alumino-silicate minerals but was not seen in the aluminum to silicon ratio. Thus, homogenization during long-range transport was suspected. In the period of continuous observations (July 3–24 of 2000), the DRUM data revealed four cases of the significantly increased atmospheric dust content level (5, 10, 15–16, and 21 of July), which correlated with the observations of the aerosol optical depth. The "bursts" of the near surface dust concentration not accompanied by changes in AOD were also observed on July 13 and 24. The mineral composition of dust was dominated by illite.

Reid and Maring[36] have characterized the main tasks and the most significant results obtained in the Puerto Rico Dust Experiment (PRIDE) in the period of intense observations on June 28 – July 24 of 2001 in order to perform the combined investigation of the long-range transport of the atmospheric dust aerosol. The principal ground site for the mission was located at Cabras Island (18.21°N, 65.60°W), which hosts a small facility several hundred meters offshore of the main island of Puerto Rico. At the site a University of Miami portable laboratory was deployed with Goddard Space Flight Center instrumentation. Two research aircraft (Piper Navajo and Cessna-172; data collected from onboard a C-130 were partly used as well) and one research vessel *Chapman* operated simultaneously along with two AERONET sites. Of great importance was the use of MODIS, MISR, CERES, GOES, and AVHRR data of satellite remote aerosol sensing.

The main PRIDE tasks were to obtain the combined information about microphysical and optical properties, as well as on the chemical composition of dust aerosol, originating from Northern Africa and then undergoing the long-range transport across the Atlantic Ocean. The particular questions to be addressed in the analysis of PRIDE data were the following: 1) how adequate are the existing numerical models of long-range transport of dust; 2) how reliable is the available information about dust sources; 3) how reliable are measured properties of dust aerosol; 4) what new (from the viewpoint of understanding of dust properties) does the analysis of combined observations give; 5) what is the correlation between *in situ* observations and results of remote sensing; 6) what is the role of nonsphericity of dust aerosol particles; 7) what is the significance of the results obtained for solution of various applied problems.

As was shown by Reid with co-workers,[37] during PRIDE, six events of inflow of the Saharan dust aerosol, underwent the long-range transport across the Atlantic Ocean, were observed on: June 28–29, July 4–6, July 9–10, July 15–16, July 21, and July 23 of 2000. In these periods, the midvisible aerosol optical depth (AOD) in Puerto Rico, averaging 0.24, varied from ~0.07 as in clean marine periods to the maximum >0.5 corresponding to the dusty atmosphere. At the same time, dust AODs near the coast of Africa averaged ~0.45 with the maximum level to 0.8. The AOD values obtained in the Puerto Rico region were somewhat lower than those observed in the previous three years, which was likely caused by the influence of stronger than usual scavenging of dust particles from the atmosphere by precipitations. The dust vertical distribution was characterized by wide variability in time.

Along with the formation of the well-known Saharan Air Layer (SAL), the conditions of much less pronounced long-range transport of dust aerosol took place. The dust aerosol often reached the altitude about 5 km, and the presence of dust in the marine boundary layer (MBL) did not correlate with some "typical" dust vertical distribution profile. In particular, no systematic correlation was found between the strength of the trade inversion and dust vertical distribution. Since the formation of trade inversions is mostly determined by local conditions, this can explain the absence of correlation between SAL and inversions. The transition from the dust transport regime in MBL to that characteristic of the lower troposphere was gradual, but with sporadic fluctuations. Some cases were characterized, however, by the existence of clear dust layers over the trade inversion. Such dust layers can be seen from distances up to hundred kilometers both in the direction of the prevailing transport and in the perpendicular direction.

As a rule, no significant changes in the particle size with altitude (vertical gradients) were observed, except for the upper part of the dusty atmosphere about 300 m thick. This is also valid for the conditions when the aerosol stratification took place. Exclusion was, however, the data collected on July 4–6 of 2000, when a significant vertical gradient of the dust particle size was observed at the altitudes from the trade inversion to the maximum aerosol altitude (about 5 km).

The factors of formation of the dust vertical profile, analyzed by Reid et al.,[37] suggest that the influence of gravitational settling of particles during the long-range transport usually was not significant. This allows the assumption to be formulated that the long-range transport across the Atlantic Ocean did not change considerably the dust vertical distribution, formed over the African continent.

However, the combined influence of large-scale settling and vertical mixing due to the moist convection (in combination with differential advection at different levels) resulted in chaotic changes of the dust vertical distribution, which were observed sometimes.

Numerous investigations are devoted to the analysis of the sensitivity of aerosol optical properties to variations of aerosol microstructure, chemical composition, and particle shape. As to the shape factor, it is insignificant for such particles as hydrated sea-salt particles or those produced upon biomass burning, but it is of great importance for dust aerosol particles, when the consideration of the particle shape strongly affect the optical properties of particles and, correspondingly, the estimates of the climatic impact of aerosol. In this context, Reid with co-authors[39] compared the data on the dust microstructure obtained by different measurement means during PRIDE in order to study the microstructure of Saharan dust coming from Africa in the period since June 28 through July 24 of 2000.

The comparison revealed quite dissimilar results, especially when using optical counters and aerodynamic methods. The analysis of results obtained using different techniques showed that, in the case of electron microscopy, large systematic errors arise when determining the number density of coarse particles (4–10 μm), which gives rise to considerable uncertainty in the microstructure parameters. Disadvantage of cascade impactors like MOUDI and DRUM are losses of particles at the inlet, achieving 40%. In the case of the DRUM impactor, large particles are sometimes destroyed in the impactor.

An APS-3300 aerodynamic particle size sensor provides for reliable measurements of the particle size, but its defect is the absence of capability to distinguish dust and sea-salt aerosol particles. A feature of a FSSP-10 airborne optical counter is more than twice overestimation of dust particles (this feature is also characteristic of other counters of coarse aerosol particles).

The microstructure parameters reconstructed from the AERONET data on the sky brightness and the total extinction of the direct solar radiation upon the assumption of the spheroidal particle shape are generally reliable, but always call for a critical analysis from the viewpoint of the possible effect of invisible cirrus clouds and insufficiently reliable regularization in solving the inverse problem. None of the techniques considered provides for the adequate (as compared to observations) values of the mass extinction coefficients and the efficiency of scattering by dust aerosol, given the spherical geometry of particles. That is why the aerosol scattering appears to be underestimated roughly by 30%. Some ideas concerning the possible ways to improve the techniques for determination of the aerosol microstructure were presented.

Livingston with co-authors[23] discussed the results of airborne (21st flight) observations of AOD and column density of water vapor (CWV) obtained with a six-channel (380.1, 450.9, 525.7, 864.5, 941.9, and 1021.3 nm) NASA Ames Airborne Tracking Sun photometer (AATS-6) in June 28–July 24 of 2000 during the Puerto Rico Dust Experiment (PRIDE). In general, aerosol extinction values calculated from AATS-6 AOD measurements acquired during aircraft profiling up to 5 km above sea level (asl) reproduced the vertical structure measured by coincident aircraft *in situ* measurements of total aerosol number concentration.

The spectral behavior of the AOD within specific layers beneath the top of the aircraft profile was consistent with the attenuation of incoming solar radiation by large dust particles or by dust plus sea salt, with mean Angström wavelength exponents of ~0.20. Values of CWV calculated from profile measurements by AATS-6 at 941.9 nm and from aircraft *in situ* measurements agree within ~4% (0.13 g/cm^2). AATS-6 AOD values measured on the Cabras Island aerosol/radiation ground site and during low-altitude aircraft flights agreed within 0.004–0.030. For the same observation times, AERONET retrievals of CWV exceeded the AATS-6 values by ~21%. AATS-6 AOD values measured during low-altitude aircraft traverses over the ocean were compared with the corresponding AOD values retrieved over water from upwelling radiance measurements by the Moderate-Resolution Imaging Spectroradiometer (MODIS), Total Ozone Mapping Spectrometer (TOMS), and GOES 8 Imager satellite sensors. This comparison has led to contradictory conclusions, which determines the urgency of the further comparisons.

Ground-based sun photometer aerosol optical measurements were conducted within the framework of the Aerosol Robotic Network (AERONET) program at Erdemli (36°33′N, 34°15′E) along the Turkish coast of the northeastern Mediterranean from January 2000 to June 2001 in order to reconstruct atmospheric AOD.[20] The measurements were used to define predominant regional aerosol optical properties, with an emphasis on mineral dust intrusion events. Dust storms affecting the region primarily originate from the central Sahara in spring, the eastern Sahara in summer, and the Middle East/Arabian peninsula in fall. And then the dust underwent the long-range transport to the northeastern Mediterranean.

Summer and fall dust intrusions usually occurred at higher altitudes (above 700 hPa), whereas urban-industrial aerosols from the north over the Balkan region, Ukraine, and Anatolia were transported to the region at lower altitudes (< 850 hPa). In addition to a drastic increase in the aerosol optical depth, in some cases up to 1.8, the dust episodes were characterized by (1) a sharp drop in the Angström coefficient to values near zero, (2) a high-scattering with single-scattering albedo greater than 0.95 ± 0.03, and the real part of the refractive index around 1.51 ± 0.07 at 440 nm, both of which acquire slightly higher values at longer wavelengths,

(3) the imaginary part of the refractive index of 0.0012 ± 0.0007 at 440 nm and 0.0075 at 870 nm, and (4) an almost neutral spectral dependence of these parameters.

Dust particles possessed a bimodal size distribution with typical volume mean radii of 2.2 μm and 0.08 μm for the coarse and fine fractions, respectively, and corresponding volume concentrations of about 1.0 and 0.1 $\mu m^3\ \mu m^{-2}$ of dust particles. It was apparent that the Saharan and Middle East desert dusts differ in their absorption index values (0.0015 and 0.0005, respectively). The difference is likely a result of their contrasting mineralogy.

Using 30 days of half-hourly, high temporal resolution GOES 8 imager data and radiative transfer calculations, Wang et al.[48] retrieved the dust aerosol optical depth (AOD) over the Atlantic Ocean (14°N–26°N, 73°W–63°W) during the Puerto Rico Dust Experiment (PRIDE). AOD values were reconstructed from satellite data on the outgoing shortwave radiation (OSR) in the visible spectral region by comparing the measured OSR values with those calculated using a discrete ordinate radiative transfer model, given the aerosol microstructure and the complex refractive index equal to $1.53–0.0015i$ at 0.55 μm.

The comparison showed that GOES 8 retrieved AOD are in a good agreement with the sun photometer (SP) derived values, with linear correlation coefficient of 0.91 and 0.80 for the two sites. The GOES 8 monthly mean AOD (0.19 ± 0.13, 0.22 ± 0.12) at 0.67 μm over the two SP sites matched the monthly mean SP AOD values (0.23 ± 0.13, 0.22 ± 0.10).

The estimates of errors caused by the neglect of the nonspherical shape of particles showed that the uncertainties ($\Delta\tau$) of the GOES 8 retrieved AOD values were mainly from the uncertainties due to the imaginary part of the refractive index ($\Delta\tau = \pm 0.05$) and inadequate surface albedo [$\Delta\tau = \pm(0.02–0.04)$]. The results obtained suggest that AOD values retrieved from GOES 8 satellite data over ocean (even at low AOD) are quite reliable to follow the space–time variations of AOD fields and, consequently, the dust content in the atmosphere.

Ginoux[16] studied the effect of nonsphericity on gravitational settling of mineral dust particles, parameterized for prolate ellipsoids and Reynolds number lower than 2. The dependence obtained (reduction of settling speed due to nonsphericity) was included in the Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model to simulate dust size distribution for April 2001.

The use of two parameterization schemes showed that for particles with the diameters greater than 5 μm, the simulated size distribution was sensitive to the numerical parameterization scheme. Changing the particle shape from spherical to nonspherical with $\lambda = 2$ (λ is the ratio of ellipsoid semiaxes; the calculations were performed at $1 < \lambda < 10$), made little difference to the simulated surface concentration and size distribution except at the periphery of the dust sources. However, when very elongated particles ($\lambda = 5$) were simulated, the differences between nonspherical and spherical particles were significant. With limited *in situ* measurements reporting most frequent λ around 1.5, the overall effect on global modeling is rather negligible.

The significantly increased content of mineral particles over southwestern, western, and central Europe resulting from a strong Saharan dust outbreak in October 11–16 of 2001 was observed at 10 stations of the European Aerosol Research Lidar Network (EARLINET). For the first time, an optically dense desert dust plume over Europe was characterized coherently with high vertical resolution on a continental scale.

As was shown by Ansmann with co-workers,[6] the main layer was located above the boundary layer (above 1-km height above sea level (asl)) up to 3–5-km height, and traces of dust particles reached heights of 7–8 km. The particle optical depth typically ranged from 0.1 to 0.5 above 1-km height asl at the wavelength of 532 nm, and maximum values close to 0.8 were found over northern Germany. The lidar observations were in qualitative agreement with the values of optical depth derived from Total Ozone Mapping Spectrometer (TOMS) data. Ten-day backward trajectories clearly indicated the Sahara as the source region of the particles. Lidar-derived particle depolarization ratios, backscatter- and extinction-related Angström exponents, and extinction-to-backscatter ratios mainly ranged from 15 to 25%, –0.5 to 0.5, and 40–80 sr, respectively, within the lofted dust plumes. The simulations were found to be consistent with the network observations.

Colarco et al.[12] simulated emissions, long-range transport of Saharan dust over the Atlantic Ocean, and dust deposition for the Puerto Rico Dust Experiment (June–July, 2000) using a three-dimensional aerosol transport model driven by assimilated meteorology. The simulation showed that the timing of high aerosol optical depth (AOD) dust events at Puerto Rico was largely independent of the timing of dust emissions, implying a persistent reservoir of suspended dust particles exists over Africa and that the intensity of long-range transport of dust aerosol depends mostly on the specific atmospheric circulation.

With the Ginoux et al.[16] source model, African and Arabian dust emissions during July 2000 were estimated to be 214 Tg for particles smaller than 10 μm in radius. Nineteen percent of the emitted mass was transported west of Africa and over the North Atlantic. Twenty percent of the dust leaving Africa passed west of Puerto Rico. Calculated AOD values are sensitive to the initial microstructure of aerosol and to the adequate parameterization of the wet deposition process.

Until recently, it seemed that the factors determining the long-range transport of the Saharan dust to the Caribbean region over tropical North

Atlantic are studied quite thoroughly. The cold and humid air from the north-east to North Africa over the Mediterranean Sea undergoes strong warming over the African continent due to the turbulent heat exchange with the surface. This is followed by advection of the heated dusted and well mixed air to the west at altitudes up to 6 km. At the very beginning of the advection process, the lower boundary of the dusted layer lifts quickly due to the effect of northeastern trade winds with relatively clean and humid air. The layer of warm, dry, and dusted air, formed in this way in the lower troposphere, was called the Saharan Aerosol Layer (SAL). It lower boundary lies at the level of 850 hPa (1.5 km), while the upper one lies near 500 hPa (6 km). As SAL moves to the west, its lower boundary lifts gradually up to about 750 hPa (3 km) and the upper boundary goes down to 550 hPa (5 km). Thus, in the Caribbean region SAL is a layer about 2 km thick.

Colarco with co-workers[13] studied the nature of SAL formation and have found that this nature is more complex than supposed earlier. It turned out, in particular, that in winter the dust transport occurs at lower altitudes than those following from the classical SAL model. The Saharan aerosol near the coast of South America was found even in the atmospheric boundary layer. The data of satellite, airborne, and ground-based observations, conducted in the period of June 28–July 24 of 2000 in Puerto Rico and including five episodes of anomalously strong dusting of the atmosphere, make clearer the SAL nature. The analysis of these data showed that the "classical" SAL was observed only in the last five days of the observations. Before this, the aerosol vertical distribution was more complex and variable.

The comparison of simulated results with observations showed that the model does a reasonable job of locating the dust plume as it emerges from Africa, but transports it somewhat farther south in the western North Atlantic Ocean than is seen in satellite imagery. The model is able to simulate low-level, uniformly mixed, and elevated vertical dust layer profiles over Puerto Rico similar to observations made in PRIDE.

The simulations suggested that the variability in the dust vertical profile across the North Atlantic Ocean was most strongly associated with descent of the dust by sedimentation and downward vertical winds. Wet removal played a key role in modulating this process. Assuming the dust to be 3.5% iron by mass, the July 2000 iron deposition into the North Atlantic Ocean was estimated to be between 0.71 and 0.88 Tg, which is consistent with estimates derived from observed surface dust mass concentrations. If annual dust deposition is five times greater than July 2000 estimates, there is an accumulation of 1 m of sediment from Saharan dust over the Florida peninsula every one million years.

Continuous record of *in situ* atmospheric measurements of Saharan dust coming to Barbados as a result of the long-range transport across the Atlantic Ocean is a more than 30 years long unique series of long-term observations. Its processing revealed fluctuations of a factor of four in surface mass concentrations between the 1960s and the 1980s.

Mahowald et al.[25] tested the hypothesis that dry topographic lows (and not disturbed sources such as cultivated areas or new desert regions) are the sources of desert dust. It was found that the inclusion of a disturbed source improves simulations in many (but not all) comparisons, but does not ensure the complete explanation of the observed dynamics of the dust aerosol. The deficit of observations prevents, however, obtaining of reliable quantitative estimates.

Cattrall et al.[9] retrieved the single-scattering albedo and scattering phase function of African mineral dust at 14 wavelengths across the visible spectrum from ground-based measurements of the aerosol optical depth and the sky radiance taken in the solar principal plane. The observations were carried out in July and August of 1998 in Fort Jefferson on Dry Tortugas Island (24°37.7′ N, 82°52.5′ W) situated about 100 km west from Key West (Florida, USA) with a 512-channel spectrometer. The retrieval algorithm employed the radiative transfer equation to solve by iteration for these properties that best reproduce the observed sky radiance, and is therefore independent of particle shape. The estimated error in the retrieved single-scattering albedo was less than 0.02. The single-scattering albedo displayed a spectral shape expected of iron-bearing minerals but was much higher than climate models have assumed, indicating that wind-blown mineral dust cools Earth more than is generally believed.

Saharan dust storms are not a single powerful source of aerosol inflow into the atmosphere from the African continent through the long-range transport. Another significant source is biomass burning.

Fires in African savannas cause about two thirds of global biomass burning in savannas. Such fires emit various minor atmospheric constituents (including CO_2, CO, NO_x, SO_2, hydrocarbons, halogen-carbon compounds, oxidated organic compounds, and aerosol) into the atmosphere. In South America, fires in savannas occur mostly in the dry season (April–October), when the weather conditions are characterized by formation of stable air masses, southeastern trade winds, and the subtropical high-pressure belt. The presence of stability layers in the atmosphere near 700 and 500 hPa layers limits the vertical motion of smoke aerosol and trace gases. Pollutants, produced over South America, undergo long-range transport, giving rise to the increase of the tropospheric ozone content over the southern region of the Atlantic Ocean, and come to the region of the Indian Ocean as well.

In August–September 2000 within the SAFARI-2000, airborne observations (CONVAIR-580) were

carried out, one of whose tasks was to study the consequences of biomass burning in South Africa from the viewpoint of the related atmospheric pollution, especially, the formation of aerosol haze layers. The observation program, described by Sinha et al.,[42] included determination of the vertical profiles (in the lower troposphere) of temperature, relative humidity, sulfur dioxide (SO_2), ozone (O_3), condensation nuclei (CN), and carbon monoxide (CO), and horizontal distributions of twenty gaseous and particulate species for five regions of southern Africa during the dry biomass burning season. The regions considered were the semiarid savannas of northeast South Africa and northern Botswana, the savanna-forest mosaic of coastal Mozambique, the humid savanna of southern Zambia, and the desert of western Namibia.

The highest average concentrations of carbon dioxide (CO_2), CO, methane (CH_4), O_3, black particulate carbon, and total particulate carbon were in the Botswana and Zambia sectors (388 and 392 ppmv, 369 and 453 ppbv, 1753 and 1758 ppbv, 79 and 88 ppbv, 2.6 and 5.5 $\mu g \cdot m^{-3}$, and 13.2 and 14.3 $\mu g \cdot m^{-3}$). This was due to intense biomass burning in Zambia and surrounding regions. The South Africa sector had the highest average concentrations of SO_2, sulfate particles, and CN (5.1 ppbv, 8.3 $\mu g\ m^{-3}$, and 6400 cm^{-3}, respectively), which derived from biomass burning and electric power plants and mining operations within this sector.

Over the arid Namibia sector there were polluted layers aloft, in which average SO_2, O_3, and CO mixing ratios (1.2 ppbv, 76 ppbv, and 310 ppbv, respectively) were similar to those measured over the other stronger polluted sectors. This was due to transport of biomass smoke from regions of widespread savanna burning in southern Angola.

Average concentrations over all sectors of CO_2 (386 ± 8 ppmv), CO (261 ± 81 ppbv), SO_2 (2.5 ± 1.6 ppbv), O_3 (64 ± 13 ppbv), black particulate carbon ($2.3 \pm 1.9\ \mu g \cdot m^{-3}$), organic particulate carbon ($6.2 \pm 5.2\ \mu g \cdot m^{-3}$), total particle mass ($26.0 \pm 4.7\ \mu g \cdot m^{-3}$), and potassium particles ($0.4 \pm 0.1\ \mu g \cdot m^{-3}$) were comparable to those in polluted urban air.

Since the majority of measurements in this study were made in locations well removed from industrial sources of pollution, the high average concentrations of pollutants reflect the effects of widespread biomass burning. Occasionally, relatively thin (~0.5 km) layers of remarkably clean air were found at ~3 km above the mean sea level, sandwiched between heavily polluted air. Near the coast, stratus clouds were formed at the altitude ~1 km.

With the Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container (CARIBIC) platform, Papaspiropoulos with co-workers[34] have measured the aerosol elemental concentrations at 9–11 km altitude in the northern hemisphere and discussed the measurements from intercontinental flights over a 2-year period between Germany and Sri Lanka/ Maldives in the Indian Ocean. Aerosol samples were collected using an impactor technique and were analyzed for the concentration of 18 elements using particle-induced X-ray emission (PIXE). The particle number concentrations, ozone, and carbon monoxide concentrations were measured simultaneously, and backward trajectories of air masses were calculated for the interpretation of the aerosol elemental concentrations.

Particulate sulfur was found to be by far the most abundant element. Its upper tropospheric concentration increased, on average, by a factor of 2 from the tropics to midlatitudes, with another factor of 2 higher concentrations in the lowermost stratosphere over midlatitudes. The consideration of backward trajectories suggested contributions from crustal sources and biomass burning, but not from meteor ablation. The trajectory data indicated aerosol transport along isentropic surfaces across the tropopause from the troposphere into the stratosphere. As a result of the prolonged residence time, ageing via oxidation of sulfur dioxide in the lowermost stratosphere was found to be a likely high-altitude, strong source that, along with the downward transport of stratospheric air, could explain the vertical gradient of particulate sulfur mass concentration around the extratropical tropopause.

In general, it can be thought that aerosol sulfur comes both below (from the surface) and from the stratosphere. The concentrations of sulfur, potassium, and iron have different annual behavior, which is most pronounced for potassium with the very low concentration in July–January and maximum concentration in spring. In contrast, the less pronounced annual behavior of the sulfur concentration is characterized by the low values in July–September in tropics and subtropics, whereas the minimum concentration in tropics is observed in July–January. The annual behavior of the iron concentration is very weak.

2. Asian aerosol

Nowadays the population in Asia is about 3.4 billion (about 60% of the global population), and Asia accounts for about a half of 136 million children born in 2000. Such an intense population growth leads to the extensive pollution of the environment, including the atmosphere, where pollutants are relatively quickly transported to long distances. Since the weather conditions characteristic of the spring in the northern hemisphere are favorable for the long-range transport of air to the east, the polluted air from Western Europe and even North America comes to Asia. On the other hand, the intense convection over the Asian continent and the seasonal west-to-east transport favor the transport of air masses across the Pacific Ocean. The existence of the jet stream, which is especially intense near Japan (reaching the

speed higher than 70 m/s), determines the time of air mass transport between Asia and North America, which sometimes amounts to only few days.

Martin et al.[27] discussed results of investigation of the long-range pollution transport over the Pacific Ocean during late winter in the northern hemisphere (February 25—April 19 of 1999). The investigations were conducted within the Pacific Exploratory Mission to the Tropics (PEM T-B) as a part of NASA's Global Tropospheric Experiment (GTE). The PEM West-B experiment was carried out as an addition to the PEM-West A mission (August 14—October 6 of 1996). The data on the content of atmospheric trace gases, such as nonmethane hydrocarbons, halocarbons, and carbon monoxide were obtained with the equipment installed onboard NASA DC-8 and P-3B laboratories flying over the Pacific Ocean. Wind data from the European Centre for Medium Range Weather Forecasts (ECMWF) were used to calculate backward trajectories along the flight routes.

Results showed that some pollutants originating from the Asian continent, and even farther west, were transported across the Pacific by the middle latitude westerly winds and reached the subtropical Pacific anticyclone where they subsided and turned southwestward under the influence of the low level trade winds. The parcels ultimately reached the western Pacific near coastal New Guinea after 20—25 days. Similar ("mirror") processes of long-range transport likely show themselves in the southern hemisphere as well, especially, when products of biomass burning in South Africa and South America are transported.

Field observation experiments carried out in the past two decades in remote regions of the globe detected the presence of significant amounts of anthropogenic pollutants in the atmosphere of these regions under environmental conditions, which were considered earlier as pollution-free. This applies, in particular, to Arctic and Antarctic, as well as remote regions of the Atlantic and Pacific Oceans. Analysis of airborne data revealed large-scale pollution plumes and aerosol layers in the free atmosphere, which were formed as a result of industrial pollution and biomass burning with the following long-range transport of atmospheric pollutants.

Aerosol plumes of continental origin are often characterized by the increased concentration of condensation nuclei of various compositions (sulfates, nitrates, dust, carbonaceous and soot aerosol) with the increased fraction of the accumulation mode (particle diameter of 0.1—1.0 m) in the aerosol microstructure. The extensive PEM-Tropics (PEMT) programs of airborne observations of the atmospheric composition over the tropical Pacific Ocean were conducted in 1996 (PEMT-A) and 1999 (PEMT-B).

The analysis of observations, carried out by Moore with co-workers,[31] revealed the presence of powerful "rivers" of continental outflow propagating into the remote marine atmosphere. Under these conditions, strong perturbations of the "pristine" marine atmosphere were observed. Most plumes were encountered in the Southern Hemisphere during PEMT A, while the opposite was observed during PEMT B. The calculation of backward trajectories clearly demonstrated the presence of such sources as biomass burning, urban/industrial emissions, and in the case of Asian outflow, dust storms. Aerosol size distributions varied from one plume to another and most combustion-derived aerosol appeared to be an internal mix of a refractory soot-like constituents in a volatile matrix.

Size-resolved volatility studies suggested the presence of ammonia in the particle phase. The radiatively important single scattering albedo ($\tilde{\omega}$) ranged from approximately 0.68 (biomass burning products with no coarse particles) to 0.94 (pollution and dust) in the free troposphere (FT) to 0.98 (pollution and sea salt) within the marine boundary layer (MBL). Vertical profiles often revealed more concentrated plumes aloft, typically situated in dry air with the ambient relative humidity <40%, and much lower values of $\tilde{\omega}$ than in the underlying MBL.

The PEM-Tropics A mission was planned with the main goal to study the clean (background) atmosphere in remote tropical regions of the World Ocean, but very soon vast pollution layers were detected in the marine troposphere. They indicated significant pollution of the "background" atmosphere over ocean in the southern hemisphere, especially, in the tropical Atlantic Ocean. In particular, in the subtropics of the southern hemisphere, an extended (> 15 000 km) plume of CO meanders around the globe was observed, which spread gradually along ~20°S. Such a plume is especially pronounced in the spring during the period of biomass burning in the subtropics of both southern and northern hemispheres.

Based on the MM5 mesoscale model, which provides high-resolution, near-global synoptic reconstructions of the weather, Chatfield with co-workers[10] carried out the numerical simulation of two situations: NASA's airborne Pacific Exploratory Mission-Tropics A (PEM-Tropics A) period, September—October 1996 and the PEM-Tropics B period, March—April 1999. For PEM-Tropics A, near-source pieces of the plume were clearly seen in the Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product.

The detailed analysis of airborne observations allowed the evolution of the plume as a whole to be reconstructed. The southern plume arises in surface accumulation regions in Africa and South America. Thunderstorm-scale venting of pollutants usually lofts the plume; however, synoptic-scale lifting can produce intense outbreaks. The plume flows eastward in the subtropical jet region as a single coherent but articulated current until it is increasingly filamented by storms in the Pacific. Similar northern subtropical plume was described for the PEM-Tropics B period.

The atmosphere is known to episodically transport dust, aerosols, and gaseous pollutants from

industrialized east Asia, the Gobi desert, and Siberian forest fires to western North America. Holzer et al.[17] have analyzed the regularities of long-range transport of dust and trace gases from these regions by estimating the transit-time probability density function (PDF) and air mass transit time (G) in order to isolate (filter out) the role of transport from other factors such as source variability and chemistry.

The PDF approach, unlike typical back-trajectory analyses, captures transport due to all possible paths and accounts for both resolved advection and subgrid processes. To obtain the statistical information about the daily mean G values, a numerical model of the global atmosphere (Model of Atmospheric Transport and Chemistry (MATCH)) was used with the input meteorological information set *a priori*.

The calculated values of G quantify the climatological mass fraction of air from the source region per interval of transit times, or ages. Over the North American west coast, this fraction peaks at transit times of ~8 days in the upper troposphere (~6 days later at the surface) for the dust and pollution regions and at 12–14 days for the Siberian region. An analysis of the variability of G at a fixed transit time allowed the authors to identify transport events and to estimate their probability of occurrence. This was illustrated for transport events to the "Pacific Northwest" (PNW) region of North America, defined as (43.8°–53.3°N) × (115.3°–124.7°W). Correlations between G averaged over the PNW and the winds at any point in the atmosphere identified large-scale anomaly structures of the flow that corresponded to favorable transport to the PNW.

On the basis of direct (*in situ*) airborne and lidar measurements, and calculated backward trajectories of air masses, Matsuki et al.[28] have followed up the process of long-range transport and the evolution of properties of dust aerosol coming to the atmosphere of the East-Asian region mostly during dust storms. Airborne measurements made in the free troposphere over central Japan in 2000–2001 revealed a small-scale yet steady transport of dust in the lower-middle free troposphere (2–6 km altitude) during spring including days with no evident dust outbreak. Such dust, found as background, was observed even in summer in the regions at altitudes higher than 4 km under the influence of remaining westerly winds. From a series of lidar observations over Nagoya (35°N, 137°E), Japan noticeable changes in aerosol characteristics were obtained in the free troposphere from spring to summer. Taklamakan desert was suggested as possible important source of the background dust.

It is for this reason that in recent years the great attention has been attracted to the problem of long-range transport of dust aerosol emitted into the atmosphere in deserts during dust storms. The influence of this process can be seen not only on the regional, but also on the global scales. It manifests itself in dust-driven changes in cloud formation and properties, ocean "fertilization" (due to the inflow of iron compounds), worsening of visibility conditions, transport of pathogens, and stimulation of respiratory diseases. The direct and indirect effect of dust on the radiative forcing determines significant (but still poorly understood) impact of dust aerosol on climate.

The most powerful sources of dust are Sahara and deserts of Central and Eastern Asia. In the last case, the Taklamakan (western China) and Gobi (north-eastern China and Mongolia) Deserts are the main contributors to the formation of natural dust aerosol.

Extremely strong dust storms occurred on the Asian continent on April 6–9 of 2001. The dust front reached the Korean Peninsula on April 8 and Japan on April 9. The atmosphere over the vast territory of the Pacific Ocean appeared under the effect of dust. By April 12–13, a part of the giant dust plume reached the USA, and later on (April 19–20) it was detected in the atmosphere over the Atlantic Ocean. Based on a comprehensive dust aerosol model developed and fully coupled to the U.S. Navy's operational Coupled Ocean/Atmospheric Mesoscale Prediction System (COAMPS) (an interactive 46-layer model for the atmosphere–ocean system with the horizontal resolution of 27 km), Liu et al.[22] simulated the dynamics of Asian dust storms for the period of April 5–15, 2001. In this case, dust was primarily generated in the Gobi and Taklamakan Deserts. The model performance was verified with satellite products, demonstrating the spatial structure of long-range dust transport, and by observations of PM10 (particles with diameter less than 10 m) and lidar data from Lanzhou, Beijing, Hefei, Tsukuba, and Nagasaki.

The model simulated the right timing and strength of dust events, predicting the following long-range dust transport. Numerical analysis showed that the first Mongolia cyclone on the 6 and 7 April and the cold front on 8 and 9 April, accompanied by a second Mongolia low, formed the major dynamic forcing patterns that mobilized, transported, and vertically redistributed the dust. Both cyclones entrained the dust and transported dust to altitudes of 8–9 km, while at the top of the cyclone, transport was anticyclonic and to the northeast.

Mechanical and convective turbulence played the major role in mixing dust upward to the top of the planetary boundary layer. In the cyclone area, vertical advection by the model-resolved upward motion in the cyclones was the dominant dynamic process that transported dust to high altitudes and to the air mass coming from the west thus entraining it into the long-range transport. The mass budget calculation for the entire simulation period revealed that about 75% (643 million ton particles with diameters < 36 μm) of the total dust production was re-deposited to the Asian deserts, 20% fell onto nondesert areas through dry and wet deposition, 1.6% fell into the China and Japan Seas, and 3.6% crossed the eastern and north-eastern boundaries of the model.

The Intensive Field Phase of the ACE-Asia field observational experiment has been carried out since

March 31 through May 4 of 2001 and focused in the region of the Yellow Sea and the Sea of Japan. Using the Georgia Tech/Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model and the meteorological forecast fields from the Goddard Earth Observing System Data Assimilation System (GEOS DAS), Chin et al.[11] forecasted the aerosol situation during the ACE-Asia field experiment. The model forecast skill was verified by comparing model-predicted data with those measured from onboard a C-130 aircraft.

The GEOS DAS meteorological forecast system showed excellent skills in predicting winds, relative humidity, and temperature. The model is also skillful in forecasting the 3D field of dust concentration, including the dust outbreak events and their trans-Pacific transport, but it constantly missed the high dust concentrations observed in the boundary layer. This can be attributed to desertification regions in the Inner Mongolia Province in China, which have developed in recent years but were not included in the model during forecasting. After incorporating the desertification sources, the model was able to reproduce the observed boundary-layer high dust concentrations over the Yellow Sea. The global model considered can not only account for the large-scale intercontinental transport, but also produce the small-scale spatial and temporal variations that are adequate for aircraft measurements planning.

From aerosol samples collected at Adak Island, Alaska, one of the southernmost Aleutian Islands, and the Poker Flat Research Range, 30 km northeast of Fairbanks, Alaska, Cahill[8] obtained the information about the aerosol microstructure and chemical composition (42 elements between sodium and lead using synchrotron X-ray fluorescence). The results of these elemental analyses and back trajectory calculations clearly demonstrated the transport of Asian aerosols into Alaska and the sub-Arctic. In addition, the aerosols at Adak Island demonstrated that northern Europe and Russia are contributing to the aerosol as well. The peak concentration of large (1.15 to 2.5 μm in aerodynamic diameter) soil aerosols at Poker Flat is 15% of that observed at Adak Island.

If it is assumed that the same air mass is sampled at both sites and that deposition is the only aerosol removal mechanism, then this implies a large deposition of crustal elements to the marine and terrestrial ecosystems along the transport path. However, the observed concentration difference could be influenced by differences in transport path, the layered nature of Asian aerosol transport, and different vertical mixing properties over Adak Island and Poker Flat. Future work is necessary to examine the relative impacts of the above processes on the observed soil element concentrations.

Uno et al.[47] described the Chemical Weather Forecast System (CFORS) developed to aid in the planning of field experiments and in the interpretation/postanalysis of observed data. The system incorporates a regional chemical transport model. This model provides for the reconstruction of 3D concentration fields of trace gases and aerosol with the high (about 90 s) time resolution. The model is used to help in interpreting the Variability of Maritime Aerosol Properties (VMAP) surface observation data.

To analyze the transport and transformation of trace gases and aerosol with the allowance for both the anthropogenic pollutants (sulfate, black carbon, carbon, CO) and the natural components (including radon and dust aerosol), the evolution of the concentration fields of trace gases and aerosol was considered. The analysis revealed the important role of weather variations on synoptic scales as a factor of pollution transport on continental scales in spring in Eastern Asia. In Ref. 47, the complex spatiotemporal structure of pollutant fields during emissions from the Asian continent to the territory of the Pacific Ocean is discussed in detail. The results of numerical simulation are in a good agreement with airborne observations.

On a network of surface stations situated at four Japanese islands (Rishiri, Sado, Hachijo, and Chichi-jima), ranging widely in latitudes from 25°N to 45°N along with the line of 140°E in the western Pacific, Matsumoto with co-workers[29] carried out regular observations of various characteristics of atmospheric aerosol, including the content of elemental carbon (EC). Reference 29 discusses the observations of EC content in the period from March to May 2001, coordinated with the Asia-Pacific Regional Aerosol Characterization Experiment (ACE-Asia) campaign. The mean concentrations of particulate EC in the four islands during this period ranged from 0.18 to 0.60 $\mu g \cdot m^{-3}$. It is interesting to note that the temporal variations in the concentrations of particulate EC in Rishiri and Sado, which are located in the northern region of Japan (35°N–45°N), synchronized with each other. The EC concentrations in Hachijo and Chichi-jima, located in the southern region of Japan (25°N–35°N), also showed similar temporal variations with each other but did not synchronize with those in Rishiri and Sado. These results demonstrate that the transport patterns of the polluted air masses to the northern regions are often different from those to the southern regions. It was also found that continental air masses from Asia are usually accompanied by pollutants over the southern regions but not over the northern regions of the northwestern Pacific.

Uno et al.[46] performed a numerical simulation of long-range transport and transformation of aerosol using the regional-scale aerosol transport model (CFORS) in order to reproduce black carbon (BC) observed within the VAMAP and APEX programs at five remote Japanese islands during the ACE-Asia experiment in spring 2001. The BC aerosol was assumed to be fully fine, which allows avoiding the consideration of gravitational sedimentation, wet deposition, and chemical ageing of carbon particles.

The dry deposition level was set as amounting to 10^{-4} (10^{-3}) m/s onto the ocean (land) surface.

Reference 46 discusses the results of two numerical experiments: (1) a control run that includes all the BC emission, and (2) a sensitivity run without open biomass burning emissions to clarify the impact of biomass burning on the BC levels in the western Pacific (the center of this region is located at a point with coordinates 25°N, 115°E; the calculations were performed on a grid with the step of 80 km for the atmospheric thickness of 0–23 km with regard for 23 nonuniformly distributed layers).

The regional aerosol model (CFORS) was shown to accurately reproduce many of the important features observed in the spatial and temporal distributions of black carbon under different atmospheric circulation regimes. At the northern stations (Rishiri and Sado), elevated BC concentrations were calculated to be mainly below the heights of 2000 m, and the biomass burning fraction was estimated to be below 20%. At the southern sites (e.g., Chichi-jima) the contribution due to biomass burning reached 32% at the surface and 52% in the free atmosphere. CFORS results indicate that the major black carbon source and transport height are different between the northern and southern sites.

Takemura et al.[45] applied the combined three-dimensional aerosol transport–radiation model, SPRINTARS, to simulate the long-range transport of the large-scale Asian dust storms from East Asia to North America during the springtime of 2001 and 2002. It was found from the calculated dust optical thickness that 10 to 20% of the Asian dust around Japan reached North America. The aerosol content is characterized by the contributions of not only dust, but also of the anthropogenic carbon and sulfate aerosol, whose AOD is comparable with AOD of the dust aerosol.

During the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia), atmospheric submicron particles were collected on Teflon filters aboard the National Center for Atmospheric Research C-130 aircraft near Japan. Particle-phase organic carbon (OC) was quantified using Fourier transform infrared (FTIR) transmission spectroscopy. Silicate, carbonate, alkane, alkene, aromatic, alcohol, carbonyl, amine, and organosulfate functional groups were identified using the spectral analysis. X-ray fluorescence identified elemental composition. The mass concentration of OC varied from 0.4 to 14.2 $\mu g \cdot m^{-3}$, and organic mass varied from 0.6 to 19.6 $\mu g \cdot m^{-3}$, representing on average 36% of the identified submicron aerosol mass.

Maria et al.[26] considered the measured carbon monoxide (CO) to OC slopes, which revealed 10 groups of air from regions described by an Asian emissions inventory. The CO/OC slope was used to compare sources and their influence on organic composition. Fifty-two percent of ACE-Asia samples had CO/OC slopes indicative of biomass burning. It was demonstrated that aerosol compositions downwind of large Asian aerosol sources show clear regional composition signatures, such as, for example, the increased concentration of dust aerosol in air masses from the Taklamakan Desert, the increased content of nitrates and ammonium sulfate in air coming from Shanghai, and the increased content of sulfates in air masses from Hokkaido.

Intensive surface measurements of the chemical composition, microstructure, and transport of atmospheric aerosols on Rishiri Island, near the northern tip of Japan (45.07°N, 141.12°E), were conducted from March to May 2001 in the period of ACE-Asia. According to the observations, the mean concentrations of nss-SO_4^{2-}, NO_3^-, NH_4^+, and nss-Ca^{2+} in aerosols were 2.48, 0.64, 0.72, and 0.17 $\mu g \cdot m^{-3}$. Elemental carbon and organic carbon in fine particles ($d < 2.5$ μm) yielded mean concentrations of 0.25 and 0.80 $\mu g \cdot m^{-3}$, respectively.

The concentrations of these species frequently increased to higher values because of outbreaks of continental polluted air masses from Asia to the Pacific, whereas under background conditions, they decreased to lower values similar to those observed over the remote regions of the ocean. The results obtained by Matsumoto et al.[30] indicate the presence of nss-SO_4^{2-} and NH_4^+ in the fine aerosol and NO_3^- and nss-Ca^{2+} in the coarse aerosol fraction.

Continentally derived NO_3^- ions are transported as coarse particles. It was often found that anthropogenic fine particles containing abundant nss-SO_4^{2-} appeared first and then were followed by large mineral particles that had absorbed NO_3^-. Short-term intrusion of the air masses containing abundant particulate carbonaceous compounds, probably due to the influence of biomass burning, also often occurred during the outflow events of continental air masses. Atmospheric behaviors of sulfate, nitrate, and carbonaceous species are different from one another, although they are all derived mainly from combustion processes (engine functioning).

Osada et al.[33] measured the number-size distribution of atmospheric aerosol particles and O_3 at Murododaira (36.6N, 137.6E, 2450 m above sea level (asl)) on the western slope of Mt. Tateyama in central Japan from January 1999 to November 2002. They considered nighttime data from 24:00 to 05:00 hours (local time). The O_3 concentration showed small variability (standard deviation of 4 ppbv) with the mean value of 40 ppbv in winter (October to February), large variability (8 ppbv) with the higher mean value of 51 ppbv in spring (March to May), and large variability (14 ppbv) with the lower mean value of 32 ppbv in summer (June to September). Highest monthly mean volume concentration (2.7 $\mu m^3/cm^3$) of the accumulation fraction particles (0.3 μm < d < 1.0 μm) was observed in June, while the mean value in winter (October to February) was 0.7 $\mu m^3/cm^3$.

On the basis of statistical analysis of the backward air mass trajectories, a stagnant airflow in summer over the coastal areas of the Yellow Sea and near Japan was inferred to be a suitable meteorological condition to form enhanced volume concentration of accumulation particles during the transport. SO_2 emission from Miyakejima volcano since August 2000 was also an important source of the summer enhancement of the accumulation fraction of aerosol. Highest monthly mean volume concentration (11.2 $\mu m^3/cm^3$) of coarse particles ($d > 1.0$ μm) was found in April. Variability of daily nighttime volume concentrations of the coarse particles was high (standard deviation of 13.6 $\mu m^3/cm^3$) in spring and low (about 2 $\mu m^3/cm^3$) in the rest of the year.

High volume concentration with large variability of the coarse particles in spring was caused by frequent arrival of Kosa (yellow dust) particles from the Asian continent. Rapid enhancement of the coarse aerosol volume concentration was often observed to increase as much as 30 times within 3 hours during Kosa phenomena. The year 2001 had particularly strong Kosa activity with a prolonged season starting early in January and ending early in July.

Schmid with co-workers[41] performed the so-called "closure" comparison between solar radiation attenuation at 550 nm by aerosols, characterized by the extinction coefficient σ_{ep}, measured with an airborne Sun photometer and derived from airborne *in situ* and ship-based lidar measurements during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia). The airborne data presented were obtained aboard the Twin Otter aircraft with the 14-channel AATS-14 sun photometer. In the period since March 31 through May 1 of 2001 within ACE-Asia, 19 flights over Japan at altitudes up to 3.8 km from Hiroshima (34.15°N, 132.23°E) were conducted.

Comparison of the vertical profiles of aerosol extinction coefficient σ_{ep} observed and calculated by four different techniques showed a good agreement with some altitude-dependent deviations in the absolute values of the aerosol extinction. The values of σ_{ep} computed from airborne *in situ* size distribution and composition measurements showed a good agreement with AATS-16 data in the marine boundary layer, but were considerably lower in layers dominated by dust if the particles were assumed to be spherical. The σ_{ep} values from airborne *in situ* scattering and absorption measurements were about 13% lower than those obtained from airborne Sun photometer for 14 vertical profiles.

Combined analysis of the lidar and airborne Sun photometer measurements revealed the prevalence of dust layers at altitudes up to 10 km with layer aerosol optical depth (from 3.5 to 10 km altitude) of 0.1 to 0.2 (500 nm) and extinction-to-backscatter ratios of 59–71 sr (523 nm). The airborne Sun photometer aboard the Twin Otter revealed a relatively dry atmosphere during ACE-Asia with the water vapor column density <1.5 cm and water vapor densities $\rho_w < 12$ g/m^3. Comparison of the layer water vapor amounts and ρ_w from the airborne Sun photometer to the same quantities measured with aircraft *in situ* sensors led to a high correlation ($r^2 = 0.96$), but the Sun photometer tended to underestimate ρ_w by 7%.

Using the NOAA14-AVHRR data and the NCEP-NCAR reanalysis data, Rajeev et al.[35a] have studied the regional distribution and long-range transport of aerosols over the Arabian Sea, Bay of Bengal and Indian Ocean during the Asian dry period (November–April) and the southwest monsoon season (June–September). The initial information was the aerosol optical depth retrieved from the AVHRR channel 1 (630 ± 50 nm) data for the territory within 25°S– 25°N, 40°E– 100°E.

Because of the contrasting air mass types, the AOD values were essentially different during the Asian dry period compared to that during the summer monsoon season. Strikingly, the AOD fields (and, thus, the aerosol content) are under the influence of the Indian subcontinent throughout the year, despite the large annual migration of ITCZ and the presence of the vast oceanic areas to the south of the continental areas.

The study demonstrated the transport of aerosols from the continental areas in the northern hemisphere to the oceanic region during the Asian dry season and a much larger-scale aerosol transport from the Arabian desert region to the Arabian Sea during the summer monsoon period. Bay of Bengal and the tropical Indian Ocean are significantly influenced by the aerosol transport from the Indian subcontinent and southeast Asia. However, the continental aerosol transport remains more or less confined to the Arabian Sea, Bay of Bengal, and northern hemisphere Indian Ocean. Throughout the year, spatial gradient in AOD is large over Arabian Sea and Bay of Bengal, particularly in the meridional direction across ~5°N to 10°N. The El Niño related forest fires in the Indonesian region, a substantially large aerosol plume, with AOD exceeding 1.0, was observed over the tropical Indian Ocean during September–November 1997.

The high level of uncertainty in the estimates of the direct and indirect aerosol impact on climate stimulated the growing interest in the analysis of processes, determining the aerosol and cloud impact on the climate formation. The uncertainties discussed are mostly attributed to the processes of complex aerosol–cloud interactions, depending on the microstructure, chemical composition, and type of clouds. The problem is even more complicated by the variability of the properties of aerosol particles due to their interaction with cloud droplets inside clouds.

The interactions between aerosol and clouds manifest themselves in two ways: through the functioning of aerosol particles as cloud condensation nuclei and through the inverse effect of clouds on aerosol, leading to changes in the number

concentration, microstructure, and chemical composition of aerosol. The properties of aerosol particles change in the process of coagulation of cloud droplets during the cloud growth due to additional matter formed as a result of oxidation of such trace gases as SO_2 in the liquid phase (in droplets), as well as through the change in the atmospheric aerosol concentration due to scavenging of particles from the atmosphere and phoretic processes.

To understand the mechanism of aerosol impact on climate, it is necessary to have the information about the aerosol concentration, properties, and lifetime under various atmospheric conditions. In this context, Levin et al.[21] investigated nucleation scavenging of aerosols by cloud drops during the passage of a shallow cold front at a mountain station situated in northern Israel on the top of Mount Meron (35.41°E, 32.99°N, 1200 m above the sea level) in the period of December 8–10 of 2000. Analysis of the air mass trajectories (long-range transport) showed that, prior to the frontal passage, the air originated from the north brought pollution particles from sources in the Eastern Europe. Following the frontal passage, the air originated from the east brought some mineral dust particles.

The results showed that sulfate, nitrate, and ammonium were the dominant compounds in the particles. Of the total sulfate-containing particles, 65% were nucleated cloud drops, nucleation scavenging of aerosols was found to be correlated with the size of the aerosol particles. Particles smaller than 0.14 μm were not significantly affected by nucleation scavenging, while the number concentration of particles larger than 0.14 μm decreased in correspondence to the increase in droplet concentrations. Under the overcast conditions, 80% of the particles in the size range 0.3–1 μm were scavenged. The concentrations of the particles with diameter smaller than 1 μm returned to their original values after the cloud dissipated.

Based on the measurement data obtained during the First International Satellite Cloud Climatology Project (ISCCP) Regional Experiment/Arctic Clouds Experiment (FIRE/ACE) flights over the Arctic Ocean, Wylie and Hudson[49] investigated variations in cloud condensation nuclei (CCN) concentration vertical profiles using back trajectories and satellite photos as additional information. Analysis of all the data showed that extended stratified clouds in the Arctic boundary layer cause a decrease of the CCN concentration in this layer due to scavenging.

The decreased concentration was observed in all cases, when air passed through clouds, except for the case of cirrus clouds, whose influence on CCN varies widely. The process of scavenging of CN and CCN by clouds leads to a decrease in their concentration at any supersaturation level. The earlier observed strong increase of the CCN concentration in the atmosphere over the Atlantic and Pacific Oceans upon the inflow of air masses from the continents was not observed over the Arctic Ocean. The maximum values of the CCN concentration were observed in the cases when air resided over the Arctic Ocean no less than 6 days in the absence of interaction between air and clouds. The vertical profiles of the CCN concentration in Arctic differ considerably from those observed in other marine regions. The maximum concentrations were detected near the highest level of airborne sensing (6.5 km, near the tropopause), that is, in contrast to the ordinary situation, the CCN concentration increased with altitude.

3. Aerosol settling processes

The final stage of the aerosol cycle is its settling onto the surface. Using the Northern Aerosol Regional Climatological Model (NARCM) of the long-range transport of dust aerosol with the allowance made for transformation of its properties due to the wet and dry deposition, Zhao et al.[50] reconstructed the evolution of the dust plume, originating from Asian deserts, to the Pacific Ocean in the northern hemisphere and reaching the western coast of America. The simulations suggested that dry deposition was a dominant dust removal process near the source areas and the removal of dust particles by precipitation was the major process over the trans-Pacific transport pathway, where wet deposition exceeded dry deposition by a factor of up to 10. The Asian dust deposition from the atmosphere to the North Pacific Ocean was correlated not only with precipitation over the North Pacific but also with the dust transport patterns.

Variations of monthly Asian dust outflow were identified with the latitudinal center of transport at 38°N in March, 42°N in April, and 47°N in May. In this connection, the monthly trans-Pacific transport patterns of Asian dust in spring were characterized in detail.[50] The transport axis extended around 30°N and 40°N in March. A zonal transport pathway around 40°N was well developed in April. However, the transport in May was separated into two pathways: an eastward zonal path and a meridional path from the source regions to the northeast of the Asian continent. On the basis of the averaged dust budgets during spring 2001, it was found that the major sources of Asian dust were located in the desert regions in China and Mongolia with an estimated dust emission of 21.5 tons km^{-2} (the export was only 8.42 tons km^{-2}), and the Loess Plateau was the main sink for Asian dust on the continent, while the Pacific Ocean was the main sink region for the rest of the aerosol transported from the continent.

Gallagher et al.[15] conducted direct (fluctuation) measurements of small (0.1–0.2 μm diameter) aerosol particle fluxes over moorland and grassland in order to analyze the effect of the surface roughness on the rate of dry deposition of aerosol. In the case of grassland, measurements were conducted both before and after cutting. Analysis of the obtained data revealed their agreement with the previous similar

results, especially, when taking into account the identical conditions of atmospheric stability. In Ref. 15, an approximate technique was proposed for parameterization of the deposition velocity V_{ds} as a function of the roughness parameter z_0:

$$V_{ds}/u^* = k_1 + k_2 (-300z/L)^{2/3},$$

where $k_1 = 0.001222 \log(z_0) + 0.003906$; $k_2 = 0.009$. Here z is the measurement height, L is the Obukhov stability length, and u^* is the local friction speed. Gallagher with co-authors[15] emphasized the necessity of further observations for different types of the surface (especially, those having the roughness in the range of 0.1–1.0 m), as well as the need of generalization of the parameterization technique to the particle size of 0.5–1.0 μm.

Along with the aerosol of soil origin, the sea-salt aerosol emitted by the World Ocean into the atmosphere is a major contributor to formation of the global mass of atmospheric aerosol (the dry mass of maritime aerosol was estimated as 10^3–10^4 Tg/yr). Hoppel with co-workers[18] conducted the numerical simulations, whose results indicated that the presence of a uniform surface source of particles, such as sea-salt aerosol, the dry deposition velocity is fundamentally different than that for a source of particles inflowing through advection or as a result of vertical mixing.

In Ref. 18, a technique was proposed for calculating the deposition velocity in these two cases. The equilibrium method of deriving the sea-salt source function from an aerosol concentration, measured at a reference height, and the deposition velocity, was shown to be of little value for particles smaller than about 5 to 10 μm in radius for two reasons: (1) The time to establish equilibrium between the source and loss by dry deposition is much longer than the typical lifetime of small particles determined by precipitation scavenging. (2) It is difficult, if not impossible, to correct for the effect of synoptic-scale vertical velocities and the effect of mixing between the marine boundary layer and the free troposphere.

Therefore, a modified technique was developed for the estimation of the sea-salt aerosol source function. This technique is based on the concept of "entrainment velocity" to take into account the effect of such processes as: 1) deposition or lift due to the large-scale divergence or convergence within MBL; 2) vertical motions due to descend or extension of MBL; 3) exchange with the free troposphere through the top MBL boundary.

Conclusions

The long-range (especially, trans-oceanic) transport of aerosol determines the various aerosol impacts on the environment and climate on regional and global scales. In this context, an important place is occupied by the pollution of the high-latitudinal troposphere due to the long-range transport of anthropogenic aerosol and trace gases, which was not considered here to avoid repetitions.[2,7] The same reasons determine the brevity of consideration of the issues concerning the long-range transport of biomass burning products and anthropogenic emissions to the territory of the Indian Ocean, which were discussed in the previous parts of this review.[3,4]

Summarizing, it should be emphasized, as before, that the available information about the long-range transport is inadequate, and this causes the need in the further development of both observation systems (with particular attention to lidar and satellite remote sensing) and in the numerical simulation of the global dynamics of the atmospheric aerosol, as well.

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